

# U.S. Ozone Air Quality under Changing Climate and Anthropogenic Emissions

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We examined future ozone ( $O_3$ ) air quality in the United States (U.S.) under changing climate and anthropogenic emissions worldwide by performing global climate-chemistry simulations, utilizing various combinations of present (1990s) and future (Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) A2 2050s) climates, and present and future (2050s; IPCC SRES A2 and B1) anthropogenic emissions. The A2 climate scenario is employed here because it lies at the upper extreme of projected climate change for the 21st century. To examine the sensitivity of U.S.  $O_3$  to regional emissions increases (decreases), the IPCC SRES A2 and B1 scenarios, which have overall higher and lower  $O_3$ -precursor emissions for the U.S., respectively, have been chosen. We find that climate change, by itself, significantly worsens the severity and frequency of high- $O_3$  events ("episodes") over most locations in the U.S., with relatively small changes in average  $O_3$  air quality. These high- $O_3$  increases due to climate change alone will erode moderately the gains made under a U.S. emissions reduction scenario (e.g., B1). The effect of climate change on high- and average- $O_3$  increases with anthropogenic emissions. Insofar as average  $O_3$  air quality is concerned, changes in U.S. anthropogenic emissions will play the most important role in attaining (or not) near-term U.S.  $O_3$  air quality standards. However, policy makers must plan appropriately for  $O_3$  background increases due to projected increases in global  $CH_4$  abundance and non-U.S. anthropogenic emissions, as well as potential local enhancements that they could cause. These findings provide strong incentives for more-than-planned emissions reductions at locations that are currently  $O_3$ -nonattainment.

## 1. Introduction

Ground-level ozone ( $O_3$ ) is one of the six criteria pollutants regulated by the United States (U.S.) Environmental Protection Agency (EPA). Despite substantial progress toward  $O_3$  pollution control in the U.S. there remain nonattainment areas (<http://www.epa.gov/oar/oaqps/greenbk/map8hrnm.html>). Although changes in U.S. anthropogenic emissions will likely play the key role in future U.S.  $O_3$  air quality, the effects of changes in climate, methane ( $CH_4$ ) abundance, and non-U.S. anthropogenic emissions cannot be discounted. While the net effect of future climate change

alone is a decrease in the global tropospheric  $O_3$  due to increased absolute humidity (1–3), there could be ground-level  $O_3$  increases in some polluted regions due to increased  $NO_x$  under warmer temperatures (4), increased biogenic volatile organic compounds (VOCs) emissions (5–7), and circulation changes (8–10). Non-U.S. anthropogenic emissions affect U.S.  $O_3$  concentrations by influencing the U.S.  $O_3$  background (11) which, for example, contributes an average 15–30 ppbv to afternoon  $O_3$  mixing ratios in surface air in the eastern U.S. and 25–35 ppbv in the western U.S. (based on model simulations (12) for the summer of 1995). Thus, future increases in non-U.S. anthropogenic emissions, especially  $CH_4$  (13, 14) and nitrogen oxides ( $NO_x$ ), could increase U.S.  $O_3$  concentrations.

Germane to this study is an ongoing multigroup modeling effort sponsored by the U.S. EPA to examine how global change will impact future U.S. air quality (7, 10, 15–17). Kunkel et al. (15) report summertime "daily maximum 8-hour  $O_3$ " (MDA8- $O_3$ ) mixing ratio increases (2100 minus 2000) of 10–24% (Intergovernmental Panel on Climate Change (IPCC) A1FI scenario) and decreases of up to 10% (IPCC B1 scenario) in the northeast U.S. due to climate change alone. On the other hand, Tagaris et al. (16) observe summertime U.S.  $O_3$  decreases (2050 minus 2000; IPCC A1B scenario) of 11–28% due to changes in climate and anthropogenic emissions, with little contribution from climate change, by itself. For the same scenario and time frame, Wu et al. (10) find summertime U.S. MDA8- $O_3$  decrease 2–15 ppbv, but with positive offsets of 2–5 ppbv in the midwest and northeast U.S. due to climate change. Their modeling suggests that future  $O_3$  episodes, especially in the midwest and northeast U.S., are far more affected by climate change than average  $O_3$ , as a result of reduced ventilation from convective and frontal passages. There is some disagreement among these modeling studies over future  $O_3$  change in the southeast U.S., with some (10) finding little change and others (7) finding relatively large increases; these disagreements appear at least in part to stem from differing treatments of isoprene chemistry (10, 18).

While the aforementioned studies are certainly relevant from a scientific standpoint, the policy maker is left with conflicting recommendations. For example, some studies find that anthropogenic emissions changes (U.S. and worldwide) will influence future U.S.  $O_3$  air quality the most (16), others find that climate change will play an equally—if not more—influential role (5, 10); others point to reductions in  $CH_4$  emissions as a cost-effective method of achieving both global (and U.S.)  $O_3$  air quality and climate objectives (14). Thus, there is a need for analyses that consider the multitude of factors influencing future U.S. air quality under a common framework. This is easier said than done, however, because a study that truly addresses all these questions under a common framework must perturb and analyze a finite but large set of parameters. Therefore, in studies of this kind, one has no choice but resort to simpler experimental designs, wherein a smaller set of parameters, those thought to be more influential than others, are varied one simulation at a time, and the results analyzed assuming linear additivity of effects.

This modeling study is a step in that direction. Since we have previously analyzed in some detail the  $O_3$  effects of climate change under present-day emissions at global and regional (U.S.) scales (see; Racherla and Adams (3, 7); hereafter referred to as RA2006 and RA2008, respectively), we focus here on the combined effects of changes in climate and anthropogenic emissions worldwide. We do not perform a detailed scenario analysis of the  $O_3$  effects of  $CH_4$  and non-

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**TABLE 1. Summary of the Simulations Performed. in Each Case, The First 6 Months Are Considered As Model Initialization**

	name	climate	anthropogenic emissions		CH <sub>4</sub> mixing ratio	duration
			U.S.	non-U.S.		
1	pc_pe	1990s	1990s	1990s	1990s	10.5 years
2	fc_pe	A2 2050s	1990s	1990s	1990s	10.5 years
3	pc_a2e	1990s	A2 2050s	A2 2050s	A2 2050s	10.5 years
4	fc_a2e	A2 2050s	A2 2050s	A2 2050s	A2 2050s	10.5 years
5	pc_b1e	1990s	B1 2050s	B1 2050s	B1 2050s	10.5 years
6	fc_b1e	A2 2050s	B1 2050s	B1 2050s	B1 2050s	10.5 years
7	pc_a2-ch4	1990s	1990s	1990s	A2 2050s	1.5 years
8	pc_a2e-non_usa	1990s	1990s	A2 2050s	1990s	1.5 years

U.S. emissions, but focus instead on quantifying their role in a high-emissions scenario.

## 2. Materials and Methods

We utilize a “unified” model of global climate (GISS GCM II-prime), gas-phase chemistry (Harvard tropospheric O<sub>3</sub>-NO<sub>x</sub>-hydrocarbon model), and aerosols including sulfate, nitrate, ammonium, black carbon, primary and secondary organic aerosol, mineral dust, and sea salt. A detailed description of the model is provided in Liao et al. (19, 20) and references therein. An atmosphere-only version of the GISS GCM II-prime, with a horizontal resolution of 4° latitude by 5° longitude and nine vertical layers extending from the surface to approximately 10 hPa is employed in this study. The first three model layers, centered at 959, 894, and 786 hPa, respectively, lie within the planetary boundary layer (PBL). Updates to the model version referenced above are provided in RA2006 (3) and RA2008 (7). As discussed in RA2008 (7), when compared against summertime (June–August) data from the U.S. EPA’s Aerometric Information Retrieval System for the years 1993–2000, the model simulates within ±5 ppbv the average MDA8-O<sub>3</sub> mixing ratios in the northeast and midatlantic states, a region in which the observed MDA8-O<sub>3</sub> is generally the highest. The discrepancies between the modeled and observed average MDA8-O<sub>3</sub> mixing ratios are most significant in a few southern states (over predicts by 10–20 ppbv), wherein the natural isoprene emissions are high, and the midwestern states (under predicts by 10–20 ppbv).

A total of eight model simulations were performed with the global model (see Table 1). Simulations 1–6, 10.5 years long each, examine the combined O<sub>3</sub> effects of changes in climate and anthropogenic emissions worldwide. We use two different climate scenarios (1990s and IPCC SRES A2 2050s) and three different anthropogenic emissions scenarios (1990s, IPCC SRES A2 2050s, and IPCC SRES B1 2050s) in these simulations. The A2 climate scenario is employed here because it lies at the upper extreme of projected climate change for the 21st century (21), although, it must be noted that the midcentury interscenario divergence is narrower compared to the year 2100. A present (1990s) or future (A2 2050s) climate scenario is imposed by changing the ocean boundary conditions (OBCs, i.e., sea surface temperature, sea-ice ratio, and mass of sea-ice) that drive the GCM. Further details on the 1990s and A2 2050s OBC data sets used here are in RA2006 (3). To examine the sensitivity of U.S. O<sub>3</sub> to regional emissions increases (decreases), the IPCC SRES A2 and B1 scenarios, which have overall higher and lower O<sub>3</sub>-precursor emissions for the United States, respectively, have been chosen. Sensitivity simulations pc\_a2-ch4 and pc\_a2e-non\_usa, 1.5 years long each, examine the U.S. O<sub>3</sub> contributions of increased CH<sub>4</sub> mixing ratio and non-U.S. anthropogenic emissions in the simulations utilizing the A2 emissions scenario, i.e., pc\_a2e and fc\_a2e. The pc\_a2-ch4 simulation corresponds to present-day climate and emis-

**TABLE 2. Annual and Global Anthropogenic O<sub>3</sub> Precursor Emissions and Methane Mixing Ratio Corresponding to the 1990s, IPCC SRES A2 2050s, and IPCC SRES B1 2050s Utilized in the Current Study<sup>a</sup>**

species	1990s	A2 2050s	B1 2050s
CH <sub>4</sub> mixing ratio (ppmv)	1.7	2.5	1.8
aircraft NO <sub>x</sub> (Tg N yr <sup>-1</sup> )	0.5	1.1	1.1
NO <sub>x</sub> (Tg N yr <sup>-1</sup> )	31.5 (20.0)	50.1 (38.0)	30.6 (18.5)
CO (Tg yr <sup>-1</sup> )	1031.0 (391.0)	1139.0 (500.0)	862.0 (222.0)
NMVOCS (Tg C yr <sup>-1</sup> )	55.4 (17.5)	84.0 (27.0)	42.5 (14.1)

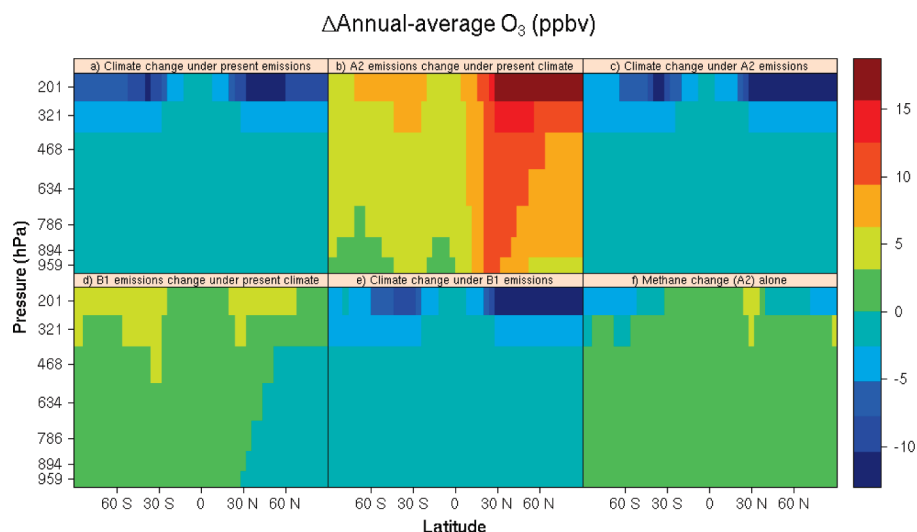
<sup>a</sup> Where applicable, values shown inside the parenthesis refer to the emissions from fossil fuel combustion and industrial sources alone, which we have changed in the 2050s. The nonmethane VOCs (NMVOCS) emitted in the model are lumped together as ≥ C<sub>4</sub> alkanes, ≥ C<sub>3</sub> alkenes, ethane, propane, and acetone.

**TABLE 3. Anthropogenic Emissions (Fossil Fuel Combustion and Industrial Sources Only) Scale Factors Implicit in the IPCC SRES A2 and B1 scenarios (2050s) for the United States**

Species	A2 2050s	B1 2050s
NO <sub>x</sub>	1.33	0.39
CO	0.87	0.53
NMVOCS	1.18	0.59

sions, but utilizes a global CH<sub>4</sub> concentration prescribed by the A2 emissions scenario (2050s). On the other hand, the pc\_a2e-non\_usa simulation corresponds to present-day climate with present-day anthropogenic emissions for North America and A2 2050 emissions for the rest of the world.

Table 2 provides a summary of the global annual anthropogenic O<sub>3</sub> precursor emissions and CH<sub>4</sub> mixing ratios used in this study. So as to obtain better agreement with observations (22), we employ the same present-day emissions utilized in previous model versions (19). We derived 4 × 5° resolution emissions scale factors (2050s A2/B1) for NO<sub>x</sub>, CO, and NMVOCS using the IPCC SRES global gridded data (1 × 1°; see [http://sres.ciesin.org/final\\_data.html](http://sres.ciesin.org/final_data.html)). These scale factors were then applied to our present-day anthropogenic emissions to arrive at the values shown in Table 2; the anthropogenic emissions scale factors for the U.S. are shown in Table 3. There are large, across-the-board increases in both worldwide- and U.S.-anthropogenic (except CO) emissions corresponding to the A2 emissions scenario. There are couple points worth noting about the B1 emissions scenario. One, the imposed CH<sub>4</sub> mixing ratio is 0.1 ppmv (100 ppbv) higher than the present-day value. Two, even though worldwide NO<sub>x</sub> emissions have reduced, those from China and India are still 50% higher than present-day (not



**FIGURE 1.** Differences in the zonal annual average  $O_3$  mixing ratios (ppbv) as a function of altitude (hPa) corresponding to (a) climate change under present emissions (fc\_pe minus pc\_pe); (b) A2 emissions change under present climate (pc\_a2e minus pc\_pe); (c) climate change under A2 emissions (fc\_a2e minus pc\_a2e); (d) B1 emissions change under present climate (pc\_b1e minus pc\_pe); (e) climate change under B1 emissions (fc\_b1e minus pc\_b1e); and (f) change in the global  $CH_4$  mixing ratio alone (pc\_a2-ch4 minus pc\_pe). The average pressure at the seven interior vertical levels (extending from the surface to the tropopause) of the model are displayed on the y-axis. All differences except those displayed in panel f are 10-year averages; panel f corresponds to a 1-year average difference.

shown). Note that the changes in anthropogenic emissions that we applied relate only to those from fossil fuel combustion (e.g., emissions from other major anthropogenic sources such as biomass burning remain unchanged). Climate-sensitive  $O_3$  precursor emissions include isoprene, biogenic lumped  $\geq C_3$  alkenes, biogenic acetone, lightning  $NO_x$ , and soil  $NO_x$ . The model assumes a static vegetation distribution and corresponding base isoprene emissions from the Global Emission Inventory Activity (GEIA) (23). The isoprene (other biogenic NMVOCs too) emitted in a model grid cell and a time step is a function of the leaf area, and the GCM provided 4-hly values of temperature and solar radiation (23, 24).

### 3. How Global-Scale Changes Affect Regional $O_3$ Air Quality

Figure 1 shows the predicted differences in the zonal and annual average  $O_3$  mixing ratios as a function of altitude. Regardless of the emissions regime, future climate change results in a decrease in the  $O_3$  background mixing ratio (Figure 1a, c, and e). Zonally and annually averaged, the typical decreases in the lower-mid troposphere are in the 0–2 ppbv range. As discussed in the introduction, the primary mechanism contributing to this decrease is the predicted increase in the absolute humidity levels which, in turn, causes an increased net destruction of  $O_3$  through  $O(^1D) + H_2O \rightarrow 2 OH$ . Note that the large model-predicted  $O_3$  decrease in the upper troposphere (UT) is due to more-than-average  $O_3$  destruction through  $O(^1D) + H_2O \rightarrow 2 OH$ , as a result of enhanced UT moistening with climate change.

Regional increases in anthropogenic  $O_3$  precursor emissions result in substantial increases in the midupper tropospheric  $O_3$  mixing ratios and, more generally, the  $O_3$  background (Figure 1b and d). For the A2 emissions scenario, the  $O_3$  increases range from 5–15 ppbv, while those for the B1 emissions scenario range from 0–5 ppbv in all latitude bands except the midhigh latitudes in the northern hemisphere, wherein the zonal-average emissions have decreased. The contribution of increased aircraft  $NO_x$  emissions is readily noticeable from the UT  $O_3$  increases in both the A2 and B1 emissions scenarios. When one considers together the global-scale effects of future changes in climate and anthropogenic

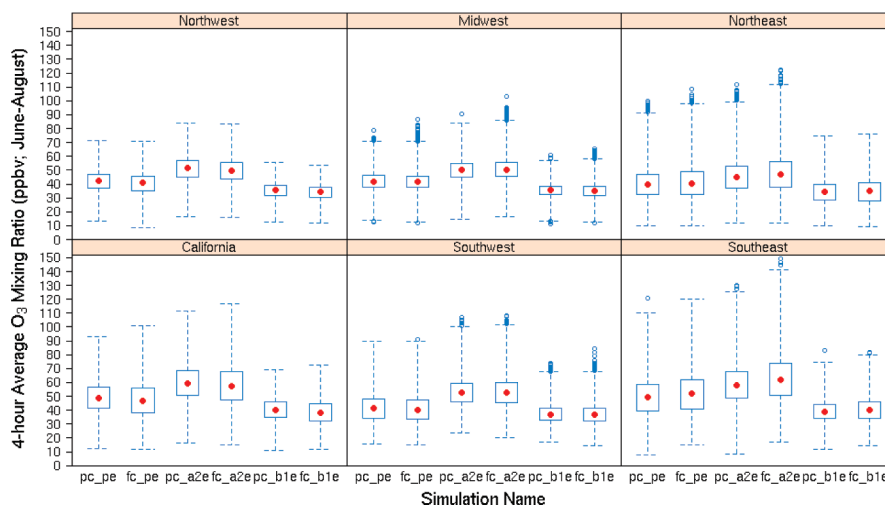
emissions, it can be seen that climate change itself alleviates somewhat the effect of increased anthropogenic emissions on the  $O_3$  background. However, as mentioned in the introduction, at the surface, climate change causes  $O_3$  increases of up to 4 ppbv in some polluted regions (not shown here; see RA2006 (3)). Also, under a pessimistic emissions scenario such as A2, up to 2 ppbv of the  $O_3$  background increase could be attributed to the increased global  $CH_4$  mixing ratio alone (Figure 1f).

### 4. Modeled Changes in U.S. $O_3$ Air Quality

In Figure 2, we examine the predicted 4 h average (no spatial averaging) surface  $O_3$  distributions (June–August) for the six contiguous regions of the U.S.: northwest, midwest, northeast, southeast, southwest, and California. There is hardly any change in the spatiotemporal averages (i.e., median values) over any of these regions due to climate change alone. This is because the two competing  $O_3$  effects of climate change—increases in locally produced  $O_3$  on one hand and decrease in the  $O_3$  “background” on the other—cancel each other out (3, 9). However, substantial increases are predicted to occur due to climate change alone at the high-end (“episodes”) of the distributions everywhere except the northwest. Readily noticeable is the increase in severity of  $O_3$  “episodes”. That the increase in climate-driven “episode” severity worsens with increasing anthropogenic emissions is also apparent (i.e., B1 < present-day < A2). In general, because the average- $O_3$  concentration and the dispersion in the predicted distributions differ considerably with the anthropogenic emissions utilized, it is not as easy to tell if the frequency of such episodic events have increased with climate change. In that sense, the increased frequency of high- $O_3$  events over the mid/southwest, wherein the average- $O_3$  and the dispersion in the distribution remain roughly constant within each climate change simulation pair, is the best evidence that climate change also increases the frequency of  $O_3$  “episodes”.

Upon closer examination of Figure 2, more subtle—but important—regional differences in the surface  $O_3$  response to climate change start to emerge. First, there is a clear

## Surface Layer of the United States



**FIGURE 2.** Box-and-whisker plots of 4 h average surface (984–934 hPa) O<sub>3</sub> mixing ratios (ppbv; June–August) for six different regions in the United States (clockwise from top left): northwest (125–115 ° W; 40–48 ° N), midwest (115–95 ° W; 36–48 ° N), northeast (95–65 ° W; 40–48 ° N), southeast (95–65 ° W; 24–40 ° N), southwest (115–95 ° W; 24–36 ° N), and California (125–115 ° W; 32–40 ° N) corresponding to simulations pc\_pe, fc\_pe, pc\_a2e, fc\_a2e, pc\_b1e, and fc\_b1e, respectively. The 4 h averaging period corresponds to the frequency at which gas-phase chemistry is integrated in the model. These data are not spatially averaged; pure ocean cells are excluded. The central box shows the data between the upper and lower quartiles (25th and 75th percentile), with the median represented by a dot; whiskers go out to data within  $\pm 3$  times the interquartile range. Values beyond that—extreme outliers—are shown as individual data points. For each simulation, the high-O<sub>3</sub> outlier concentrations are a measure of the severity of “episodes”, while the sheer number of high-O<sub>3</sub> outlier data points reflect the frequency.

west–east gradient to the high-O<sub>3</sub> sensitivity, with the west coast states being the least sensitive and the east coast states being the most sensitive. Second, among the eastern and midwestern states, the response in the southeast appears to be fundamentally different in that the entire O<sub>3</sub> distribution appears to be shifted (as a function of the anthropogenic emissions utilized, of course). This means that the O<sub>3</sub> produced locally in the southeast has increased substantially due to climate change, with little accompanying change in the average lifetime of O<sub>3</sub> itself. By comparison, only outliers at the high-end are noticeably affected in the northeast, midwest, and southwest, with the low-end and median values remaining either unchanged or decreasing slightly; while these high-O<sub>3</sub> increases could have come from a variety of mechanisms, the asymmetric changes in the distribution clearly reflect a shortening of the average O<sub>3</sub> lifetime.

Racherla and Adams (7) previously examined in detail the surface O<sub>3</sub> response to climate change over the eastern U.S. (under present-day anthropogenic emissions) and concluded that increases in the O<sub>3</sub> chemical production contributed most to the high-O<sub>3</sub> increases over the southeast and northeast. Furthermore, they identified the more than 20% annual increase in climate-sensitive isoprene emissions over the southeast and midatlantic as responsible for 50–60% of the high-O<sub>3</sub> increase there. Over the northeast, increases in NO<sub>x</sub> due to warmer temperatures and VOC-related chemical radicals (e.g., HO<sub>2</sub>·) due to increased absolute humidity were identified as leading causes for the high-O<sub>3</sub> increases. While the aforementioned RA2008 (7) study did ample justice to explaining the increased severity of high-O<sub>3</sub> events due to climate change, they presented inconclusive evidence regarding the role of changes in synoptic-scale circulations, which some previous studies (8–10) have identified will play an important role in causing more frequent O<sub>3</sub> “episodes” over the midwest and northeast under future meteorological conditions. However, given the broadly consistent high-O<sub>3</sub> response (i.e., increases) across a range of anthropogenic emissions in the mid/southwest—a region that

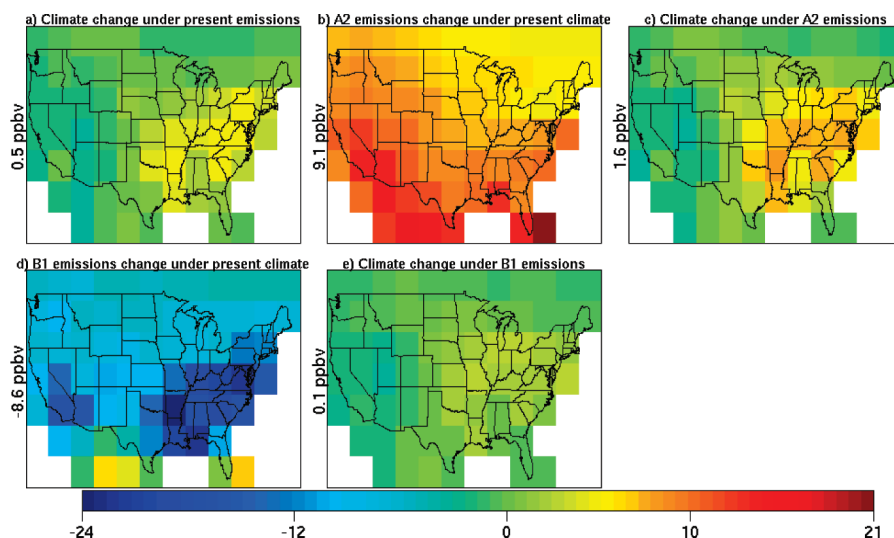
does not experience much increased O<sub>3</sub> chemical production in the model—we are beginning to think that these increases are indicative of changes in synoptic-scale circulation. Further work is necessary to confirm this, however.

The surface O<sub>3</sub> response to the imposed anthropogenic emissions changes (Figure 2) is more along expected lines. All regions except the northeast experience large spatiotemporally averaged increases and decreases of up to +10 ppbv and –10 ppbv corresponding to A2 and B1 emissions scenarios, respectively. From a policy perspective, it should be heartening to see the O<sub>3</sub> air quality benefits due to local anthropogenic emissions reductions (B1). However, for the same emissions reductions scenario, the benefits at the very low-end are practically negligible, reflecting an increase in the U.S. O<sub>3</sub> background due to increases in CH<sub>4</sub> mixing ratio (by 100 ppbv) and non-U.S. anthropogenic emissions.

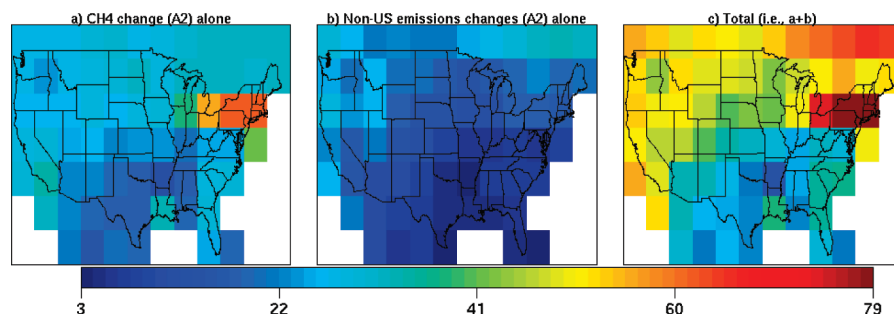
Figure 3 shows the spatial distribution of predicted differences in MDA8-O<sub>3</sub> mixing ratios in the surface layer of the U.S. averaged over June–August (10 years). Note that the U.S. EPA utilizes MDA8-O<sub>3</sub> as its O<sub>3</sub> air quality metric. The MDA8-O<sub>3</sub> changes strengthen many of the key findings thus far. More than anything else though, the predicted U.S. MDA8-O<sub>3</sub> changes reiterate that average-O<sub>3</sub> is most sensitive to anthropogenic emissions changes. However, it is hard to make further inferences at the moment because the contributions of increased CH<sub>4</sub> mixing ratio and non-U.S. anthropogenic emissions have not yet been quantified. This is examined next.

Figure 4 examines the relative contributions (%) of increases in CH<sub>4</sub> mixing ratio and non-U.S. anthropogenic emissions to the predicted surface O<sub>3</sub> response in the A2 emissions scenario. The MDA8-O<sub>3</sub> differences themselves (not shown) range from 1.4 to 4.6 ppbv (U.S. average of 2.4 ppbv) and 0.3 to 2.7 ppbv (U.S. average of 1.3 ppbv) corresponding to increased CH<sub>4</sub> mixing ratio and non-U.S. anthropogenic emissions, respectively. In terms of relative contributions to the predicted surface O<sub>3</sub> response in the A2 emissions scenario, the O<sub>3</sub> effect of increased





**FIGURE 3.** Differences in the average (June–August) MDA8-O<sub>3</sub> mixing ratios (ppbv) in the surface layer (984–934 hPa) of the United States corresponding to (a) climate change under present emissions (fc\_pe minus pc\_pe); (b) A2 emissions change under present climate (pc\_a2e minus pc\_pe); (c) climate change under A2 emissions (fc\_a2e minus pc\_a2e); (d) B1 emissions change under present climate (pc\_b1e minus pc\_pe); (e) climate change under B1 emissions (fc\_b1e minus pc\_b1e). Since we utilize a 4 h time step for the gas-phase chemistry, MDA8-O<sub>3</sub> mixing ratios are calculated by applying a running 2-value averaging window to the model output and then determining the maximum for each day. All differences correspond to 10 year averages. In each case, the mean difference is displayed to the left of the panel. White cells are pure-ocean grid cells.



**FIGURE 4.** Normalized (%) average (June–August) MDA8-O<sub>3</sub> differences in the surface layer (984–934 hPa) of the U.S. corresponding to (a) increased CH<sub>4</sub> mixing ratio alone (pc\_a2-ch4 minus pc\_pe); (b) increased non-U.S. emissions alone (pc\_a2e-non\_usa minus pc\_pe); and (c) total, i.e., = a + b. The displayed MDA8-O<sub>3</sub> differences have been normalized, cell-by-cell, by the MDA8-O<sub>3</sub> differences between the pc\_a2e and pc\_pe simulations, so as to emphasize the relative contributions of increased CH<sub>4</sub> mixing ratio and non-U.S. anthropogenic emissions to the predicted surface O<sub>3</sub> response in the A2 emissions scenario. All differences correspond to 1-year averages. White cells are pure-ocean grid cells. See the caption to Figure 3 for the MDA8-O<sub>3</sub> calculation.

CH<sub>4</sub> mixing ratio is largest (40–60% contributions) in the New England, New York, Pennsylvania, Ohio, and New Jersey model grid cells, followed by more than modest increases (20–40% contributions) in the California, Carolina (North/South), Georgia, and East Texas model grid cells. The smallest effects (<10%) are generally in the southwest/east, implying that local (U.S.) anthropogenic emissions changes dominate here.

Thus, model grid cells that encompass major urban centers (“VOC-limited” O<sub>3</sub> chemistry (4)) receive large MDA8-O<sub>3</sub> contributions from increased CH<sub>4</sub> mixing ratio, whereas those that encompass rural areas (“NO<sub>x</sub>-limited” O<sub>3</sub> chemistry) have smaller contributions. Although it seems a far stretch to suggest that a coarse 4 × 5° resolution model such as ours is capable of resolving VOC-NO<sub>x</sub> limitations accurately, other modeling studies—most notably Fiore et al. (14) have arrived at similar conclusions: “the surface ozone response to CH<sub>4</sub> is strongly enhanced in locations with NO<sub>x</sub>-saturated (i.e., VOC-limited) chemistry, and weakly enhanced in regions of downwelling air”. Note that the study used the MOZART-2 (25) tropospheric chemistry model with a horizontal resolution of 1.9 × 1.9°.

The most noticeable surface O<sub>3</sub> contributions (20–30%) from increased non-U.S. anthropogenic emissions are predicted to occur along the west coast (Figure 4b). This is due to the increased Asian outflow of O<sub>3</sub> and NO<sub>y</sub>—defined here as NO<sub>x</sub> plus its oxidation products—associated with the steep anthropogenic emissions increases in that region as prescribed by the A2 emissions scenario. As one would expect from present-day hemispheric flow patterns (11), the eastern U.S. experiences smaller surface O<sub>3</sub> contributions (≤10%) from increases in Asian anthropogenic emissions. These findings are similar to those described in Jacob et al. (26).

The combined O<sub>3</sub> effects of increased CH<sub>4</sub> mixing ratio and non-U.S. anthropogenic emissions contribute anywhere between 20 and 80% of the predicted average MDA8-O<sub>3</sub> increases in the A2 emissions scenario (Figure 4c). Although these findings are specific to the A2 emissions scenario, some broad inferences can be made for other emissions scenarios as well (e.g., B1). Specifically, increased CH<sub>4</sub> mixing ratio (by 100 ppbv) and Asian anthropogenic emissions—factors that persist in the B1 emissions scenario—potentially explain why the rest of the U.S.

experiences moderate MDA8-O<sub>3</sub> decreases compared to some southeast/midatlantic states.

## 5. Discussion

Going from the scientific findings presented here to policy is rendered difficult by many factors. First and foremost, the coarse resolution of the modeling system utilized in this study. As a result of its coarse resolution, the model generally tends to underestimate O<sub>3</sub> concentrations in urban hot spots and overestimate them over rural areas (27). These model biases generally creep into the predicted O<sub>3</sub> effects of changes in climate and emissions. On a related note, the ability of such a model—or any other global chemical transport model for that matter—to truly capture urban—regional O<sub>3</sub> “episodes” is also diminished. Despite these issues, global models continue to be used for future air quality modeling because of the prohibitive computational costs associated with performing long-term integrations of high-resolution air quality models for more than a few scenarios. Second, fundamental uncertainties regarding isoprene-O<sub>3</sub>-NO<sub>x</sub> chemistry (18) leave the predicted climate-driven O<sub>3</sub> increases over the southeast U.S. open to question. Third, there are other global-scale changes, not considered here, that could potentially affect the findings of this study. These include, but are not limited to, changes in hemispheric transport patterns (11), the stratosphere—troposphere exchange (28), and land use-vegetation changes (21). Last, but not the least, the problems associated with using broad, global-scale emissions scenarios such as the IPCC SRES for future air quality modeling must not be overlooked. Notwithstanding these shortcomings, the findings discussed previously capture some important features of U.S. O<sub>3</sub> air quality under changing climate and emissions that could, at the very least, be used as a guidepost for policy makers.

The most important policy implications of these findings are the following. One, insofar as average O<sub>3</sub> air quality is concerned, changes in U.S. anthropogenic emissions will play the most important role in attaining (or not) near-term U.S. O<sub>3</sub> air quality standards. However, policy makers must plan appropriately for O<sub>3</sub> background increases due to projected increases in global CH<sub>4</sub> abundance and non-U.S. anthropogenic emissions, as well as potential local enhancements that they could cause. Two, climate change, by itself, significantly worsens the severity and frequency of high-O<sub>3</sub> events (“episodes”) over most locations in the U.S., with relatively small changes in average O<sub>3</sub> air quality. There appears to be a general consensus among both regional and global modeling studies on this. If projected U.S. emissions reductions (e.g., IPCC SRES B1) do materialize, these high-O<sub>3</sub> increases due to future climate change alone will moderately erode the gains made. Three, the effect of climate change on high- and average-O<sub>3</sub> increases with anthropogenic emissions.

The above finding that the O<sub>3</sub> effect of climate change increases with anthropogenic emissions utilized has important regulatory implications. It implies, for example, that the benefits of anthropogenic emissions reductions under a future climate will be 2-fold. The O<sub>3</sub> decreases due to the NO<sub>x</sub>/VOC emissions reductions themselves, which is a direct benefit, and the minimized O<sub>3</sub> increase due to climate change itself, which could be viewed as an indirect benefit. Conversely, the O<sub>3</sub> increases due to anthropogenic emissions increases will be amplified by future climate change, which could be viewed as a climate “penalty” (10). These findings provide strong incentives for more-than-planned emissions reductions at locations that are currently O<sub>3</sub> -nonattainment.

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